526 Rec'd PCT/PTO 18 JUL 2001

FORM PTO-1390 REV. 2/01 — U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER: 05725.0944

U.S. APPLICATION NO. (If known, see 37 CFR 1.5): 09/889503

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/FR00/00073	January 14, 2000	January 19, 1999

TITLE OF INVENTION: NOVEL CATIONIC OXIDATION BASES, THEIR USE FOR THE OXIDATION DYEING OF KERATINOUS FIBRES, DYEING COMPOSITIONS AND METHODS OF DYEING

APPLICANT(S) FOR DO/EO/US: Eric TERRANOVA, Aziz FADLI and Alain LAGRANGE

Applicant(s) herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. X This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. X The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. X A copy of the International Application as filed (35 U.S.C. 371 (c)(2)).
   a. ☐ is attached hereto (required only if not communicated by the International Bureau).
   b. X has been communicated by the International Bureau.
   c. ☐ is not required, as the application was filed with the United States Receiving Office (RO/US).
6. X An English language translation of the International Application as filed (35 U.S.C. 371 (c)(2)).
   a. X is attached hereto.
   b. ☐ has been previously submitted under 35 U.S.C. 154 (d)(4).
7. X Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)).
   a. ☐ are attached hereto (required only if not communicated by the International Bureau).
   b. ☐ have been communicated by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. X have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. ☐ Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A Substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821-1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154 (d)(4).
19. ☐ A second copy of the English language translation of the international application 35 U.S.C. 154 (d)(4).
20. X Other items or information:
    a. X Copy of cover page of International Publication No. WO **00/43396**
    b. ☐ Copy of Notification of Missing Requirements.
    c. ☐

JC18 Rec'd PCT/PTO 18 JUL 2001

U.S. APPLICATION NO. (If known, see 37 CFR 1.5)	INTERNATIONAL APPLICATION NO. PCT/FR00/00073	ATTORNEY'S DOCKET NUMBER 05725.0944
09/889503		

21. ☒ The following fees are submitted:

	CALCULATIONS PTO USE ONLY
BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):	
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO **$1000.00**	
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO **$860.00**	860.00
International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search fee (37 CFR 1.445(a)(2)) paid to USPTO **$710.00**	
International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) **$690.00**	
International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33 (1)-(4) **$100.00**	

			ENTER APPROPRIATE BASIC FEE AMOUNT =	$860.00	
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).				$	
CLAIMS	NUMBER FILED		NUMBER EXTRA	RATE	
Total Claims	24	- 20 =	4	x $18.00	$72.00
Independent Claims	1	-3 =		x $80.00	$
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				+$270.00	$270.00
			TOTAL OF THE ABOVE CALCULATIONS =	$1202.00	
☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by ½.				$	
			SUBTOTAL =	$1202.00	
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest priority date (37 CFR 1.492(f)).				$	
			TOTAL NATIONAL FEE =	$1202.00	
Fee for recording the enclosed assignment (37 CFR 1.21 (h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). **$40.00** per property. +				$	
			TOTAL FEES ENCLOSED =	$1202.00	
				Amount to be refunded:	$
				charged:	$

a. X A check in the amount of $ 1202.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. _____ in the amount of $ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 06-0916. A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Finnegan, Henderson, Farabow, Garrett & Dunner, L.L.P
1300 I Street, N.W.
Washington, D.C. 20005-3315

SIGNATURE
Ernest F. Chapman / 25,961
NAME/REGISTRATION NO.

DATED: July 18, 2001

NOVEL CATIONIC OXIDATION BASES, THEIR USE FOR THE OXIDATION DYEING OF KERATINOUS FIBRES, DYEING COMPOSITIONS AND METHODS OF DYEING

The invention relates to novel pyrazolo[1,5-a]pyrimidines comprising at least one cationic group Z, Z being selected from quaternized aliphatic chains, aliphatic chains containing at least one quaternized saturated ring and aliphatic chains containing at least one quaternized unsaturated ring, to their use as oxidation base for the oxidation dyeing of keratinous fibres, to the dyeing compositions comprising them, and to the methods of oxidation dyeing which employ them.

It is known to dye keratinous fibres, and especially human hair, with dyeing compositions comprising oxidation dye precursors, especially ortho- or para-phenylenediamines, ortho- or para-aminophenols, and heterocyclic compounds such as diaminopyrazole derivatives, which are referred to generally as oxidation bases. The oxidation dye precursors or oxidation bases are colourless or slightly coloured compounds which, when combined with oxidizing products, have the capacity to give rise to coloured and colouring compounds by virtue of a process of oxidative condensation.

It is also known that the shades obtained with these oxidation bases may be varied by combining

them with couplers or coloration modifiers, the latter being selected in particular from aromatic meta-diamines, meta-aminophenols, meta-diphenols and certain heterocyclic compounds.

The variety of molecules employed as oxidation bases and couplers makes it possible to obtain a rich palette of colours.

The so-called permanent coloration obtained by virtue of these oxidation dyes is required, moreover, to satisfy a certain number of requirements. Hence it must have no toxicological drawbacks, must allow shades of the desired intensity to be obtained, and must have good resistance to external agents (light, inclement weather, washing, permanent-waving, perspiration and friction).

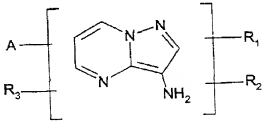
The dyes must also allow white hair to be covered and, finally, they must be as unselective as possible; in other words, they must allow the smallest possible differences in coloration to be produced over the entire length of a single keratinous fibre, which may indeed be sensitized (i.e. damaged) differently between its tip and its root.

The applicant has now discovered, completely unexpectedly and surprisingly, that new pyrazolo[1,5-a]pyrimidines of formula (I) defined below, comprising at least one cationic group Z, Z being selected from quaternized aliphatic chains, aliphatic chains containing at least one quaternized

saturated ring and aliphatic chains containing at least one quaternized unsaturated ring are not only suitable for use as oxidation dye precursors, but also make it possible to obtain dyeing compositions which lead to intense colorations in a wide palette of colours and have excellent properties of resistance to the various treatments which the keratinous fibres may undergo.

It is these discoveries which form the basis of the present invention.

The invention therefore provides, firstly, novel compounds of formula (I) below, and their addition salts with an acid:



(I)

in which:

- R_1, R_2 and R_3, which may be identical or different, represent a hydrogen atom; a halogen atom; a group Z; a (C_1-C_6 alkyl)carbonyl radical; an amino(C_1-C_6 alkyl)carbonyl radical; an N-Z-amino(C_1-C_6 alkyl)carbonyl radical; an N-(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl radical; an N,N-di(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl radical; an amino(C_1-C_6 alkyl)carbonyl(C_1-C_6 alkyl) radical; an N-Z-amino(C_1-C_6 alkyl)carbonyl(C_1-C_6 alkyl) radical; an N-(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl(C_1-C_6 alkyl)

radical; an N,N-di(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl(C_1-C_6 alkyl) radical; a carboxyl radical; a (C_1-C_6 alkyl)carboxyl radical; a (C_1-C_6 alkyl)sulphonyl radical; an aminosulphonyl radical; an N-Z-aminosulphonyl radical; an N-(C_1-C_6 alkyl)aminosulphonyl radical; an N,N-di(C_1-C_6 alkyl)-aminosulphonyl radical; an aminosulphonyl(C_1-C_6 alkyl) radical; an N-Z-aminosulphonyl(C_1-C_6 alkyl) radical; an N-(C_1-C_6 alkyl)aminosulphonyl(C_1-C_6 alkyl) radical; an N,N-di(C_1-C_6 alkyl)aminosulphonyl(C_1-C_6 alkyl) radical; a carbamyl radical; an N-(C_1-C_6 alkyl)carbamyl radical; an N,N-di(C_1-C_6 alkyl)carbamyl radical; a carbamyl(C_1-C_6 alkyl) radical; an N-(C_1-C_6 alkyl)carbamyl(C_1-C_6 alkyl) radical; an N,N-di(C_1-C_6 alkyl)carbamyl(C_1-C_6 alkyl) radical; a C_1-C_6 alkyl radical; a hydroxyl radical; a nitro radical; a C_1-C_6 monohydroxyalkyl radical; a C_2-C_6 polyhydroxyalkyl radical; a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical; a C_1-C_6 trifluoroalkyl radical; a cyano radical; a group OR_6 or SR_6; an amino radical; an N-(C_1-C_6 alkyl)amino radical; an N,N-di(C_1-C_6 alkyl)amino radical (where the two alkyl substituents may form a 5- or 6-membered ring); an N-hydroxy(C_1-C_6 alkyl)amino radical; an N,N-bis(hydroxy(C_1-C_6 alkyl))amino radical; an N-polyhydroxy(C_2-C_6 alkyl)amino radical; an N,N-bis(polyhydroxy(C_2-C_6 alkyl))amino radical; an amino(C_1-C_6 alkyl)amino radical in which the terminal amino group is unsubstituted or substituted by one or

two C_1-C_6 alkyl radicals, where the said alkyl radicals may form a saturated or unsaturated, 5- or 6-membered ring; an amino group protected by a (C_1-C_6 alkyl)carbonyl, trifluoro(C_1-C_6 alkyl)carbonyl, amino(C_1-C_6 alkyl)carbonyl, N-Z-amino(C_1-C_6 alkyl)-carbonyl, N-(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl, N,N-di(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl or formyl radical or by a group Z;

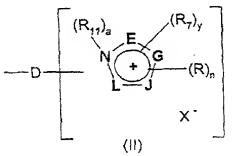
- R_6 denotes a C_1-C_6 alkyl radical; a C_1-C_6 monohydroxyalkyl radical; a C_2-C_6 polyhydroxyalkyl radical; a group Z; a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical; an aryl radical; a benzyl radical; a C_1-C_6 carboxyalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carboxyalkyl radical; a C_1-C_6 cyanoalkyl radical; a C_1-C_6 carbamylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 N,N-di(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 trifluoroalkyl radical; a C_1-C_6 aminosulphonylalkyl radical; a C_1-C_6 N-Z-aminosulphonylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)aminosulphonylalkyl radical; a C_1-C_6 N,N-di(C_1-C_6 alkyl)aminosulphonylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)sulphinylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)sulphonylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carbonylalkyl radical; a C_1-C_6 aminoalkyl radical; a C_1-C_6 aminoalkyl radical whose amine is substituted by one or two identical or different radicals selected from C_1-C_6 alkyl, monohydroxy(C_1-C_6 alkyl), polyhydroxy(C_2-C_6 alkyl), (C_1-C_6 alkyl)-

carbonyl, formyl, trifluoro(C_1-C_6 alkyl)carbonyl and (C_1-C_6 alkyl)sulphonyl radicals or by a group Z;

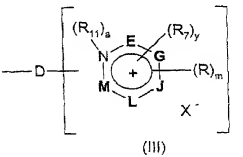
- A represents a group $-NR_4R_5$ or a hydroxyl radical;
- R_4 and R_5, which are identical or different, represent a hydrogen atom; a group Z; a C_1-C_6 alkyl radical; a C_1-C_6 monohydroxyalkyl radical; a C_2-C_6 polyhydroxyalkyl radical; a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical; an aryl radical; a benzyl radical; a C_1-C_6 cyanoalkyl radical; a C_1-C_6 carbamylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 N,N-di(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 thiocarbamylalkyl radical; a C_1-C_6 trifluoroalkyl radical; a C_1-C_6 sulphoalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carboxyalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)sulphinylalkyl radical; a C_1-C_6 aminosulphonylalkyl radical; a C_1-C_6 N-Z-aminosulphonylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)-aminosulphonylalkyl radical; a C_1-C_6 N,N-di(C_1-C_6 alkyl)aminosulphonylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carbonylalkyl radical; a C_1-C_6 aminoalkyl radical; a C_1-C_6 aminoalkyl radical whose amine is substituted by one or two identical or different radicals selected from C_1-C_6 alkyl, C_1-C_6 monohydroxyalkyl, C_2-C_6 polyhydroxyalkyl, (C_1-C_6 alkyl)carbonyl, (C_1-C_6 alkyl)sulphonyl, formyl and trifluoro(C_1-C_6 alkyl)carbonyl radicals or by a group Z;

one and only one of the radicals R_4 and R_5 may also represent a (C_1-C_6 alkyl)carbonyl; formyl; trifluoro(C_1-C_6 alkyl)carbonyl; amino(C_1-C_6 alkyl)carbonyl, N-Z-amino(C_1-C_6 alkyl)carbonyl; N-(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl; or N,N-di(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl radical;

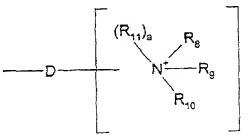
- Z is selected from the unsaturated cationic groups of formulae (II) and (III) below and the saturated cationic groups of formula (IV) below:



(II)



(III)



(IV)

in which:

- D is a linker which represents an alkyl chain containing preferably 1 to 14 carbon atoms, is linear or branched and may be interrupted by one or more heteroatoms such as oxygen, sulphur or nitrogen atoms and may be substituted by one or more hydroxyl or

C_1-C_6 alkoxy radicals, and may carry one or more ketone functions;

- the ring members E, G, J, L and M, which are identical or different, represent a carbon, oxygen, sulphur or nitrogen atom;
- n is an integer between 0 and 4, inclusively;
- m is an integer between 0 and 5, inclusively;
- the radicals R, which are identical or different, represent a group Z, a halogen atom, a hydroxyl radical, a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical, a C_2-C_6 polyhydroxyalkyl radical, a nitro radical, a cyano radical, a C_1-C_6 cyanoalkyl radical, a C_1-C_6 alkoxy radical, a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical, an amido radical, an aldehydo radical, a carboxyl radical, a C_1-C_6 alkylcarbonyl radical, a thio radical, a C_1-C_6 thioalkyl radical, a (C_1-C_6 alkyl)thio radical, an amino radical, an amino radical protected by a (C_1-C_6 alkyl)carbonyl, carbamyl or (C_1-C_6 alkyl)sulphonyl radical; a group NHR″ or NR″R‴ in which R″ and R‴, which are identical or different, represent a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical or a C_2-C_6 polyhydroxyalkyl radical;
- R_7 represents a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical, a C_2-C_6 polyhydroxyalkyl radical, a C_1-C_6 cyanoalkyl radical, a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical, a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical, a carbamyl(C_1-C_6 alkyl) radical, a C_1-C_6

(C_1-C_6 alkyl)carboxyalkyl radical, a benzyl radical, or a group Z;

- R_8, R_9 and R_{10}, which are identical or different, represent a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical, a C_2-C_6 polyhydroxyalkyl radical, a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical, a C_1-C_6 cyanoalkyl radical, an aryl radical, a benzyl radical, a C_1-C_6 amidoalkyl radical, a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical or a C_1-C_6 aminoalkyl radical whose amine is protected by a (C_1-C_6 alkyl)carbonyl, amido, carboxyl or (C_1-C_6 alkyl)sulphonyl radical; two of the radicals R_8, R_9 and R_{10} may also form, together with the nitrogen atom to which they are attached, a saturated 5- or 6-membered carbon-containing ring or one containing one or more heteroatoms such as, for example, a pyrrolidine ring, a piperidine ring, a piperazine ring or a morpholine ring, it being possible for the said ring to be unsubstituted or substituted by a halogen atom, a hydroxyl radical, a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical, a C_2-C_6 polyhydroxyalkyl radical, a nitro radical, a cyano radical, a C_1-C_6 cyanoalkyl radical, a C_1-C_6 alkoxy radical, a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical, an amido radical, an aldehydo radical, a carboxyl radical, a C_1-C_6 ketoalkyl radical, a thio radical, a C_1-C_6 thioalkyl radical, a (C_1-C_6 alkyl)thio radical, an amino radical, or an amino radical protected by a

(C_1-C_6 alkyl)carbonyl, carbamyl or (C_1-C_6 alkyl)sulphonyl radical;

one of the radicals R_8, R_9 and R_{10} may also represent a second group Z, identical to or different from the first group Z;

- R_{11} represents a C_1-C_6 alkyl radical; a C_1-C_6 monohydroxyalkyl radical; a C_2-C_6 polyhydroxyalkyl radical; an aryl radical; a benzyl radical; a C_1-C_6 aminoalkyl radical, a C_1-C_6 aminoalkyl radical whose amine is protected by a (C_1-C_6 alkyl)carbonyl, carbamyl or (C_1-C_6 alkyl)sulphonyl radical; a C_1-C_6 carboxyalkyl radical; a C_1-C_6 cyanoalkyl radical; a C_1-C_6 carbamylalkyl radical; a C_1-C_6 trifluoroalkyl radical; a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical; a C_1-C_6 sulphonamidoalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carboxyalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)-sulphinylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)sulphonyl-alkyl radical; a C_1-C_6 (C_1-C_6 alkyl)ketoalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)sulphonamidoalkyl radical;
- a and y are integers equal to 0 or 1; with the following conditions:
  - in the unsaturated cationic groups of formula (II):
    - when a = 0, the linker D is attached to the nitrogen atom,
    - when a = 1, the linker D is attached to one of the ring members E, G, J or L,
    - y can adopt the value 1 only

1) when the ring members E, G, J and L simultaneously represent a carbon atom and when the radical R_7 is carried by the nitrogen atom of the unsaturated ring; or else

2) when at least one of the ring members E, G, J and L represents a nitrogen atom to which the radical R_7 is attached;

- in the unsaturated cationic groups of formula (III):
  - when a = 0, the linker D is attached to the nitrogen atom,
  - when a = 1, the linker D is attached to one of the ring members E, G, J, L or M,
  - y can adopt the value 1 only when at least one of the ring members E, G, J, L and M represents a divalent atom and when the radical R_7 is carried by the nitrogen atom of the unsaturated ring;
- in the cationic groups of formula (IV):
  - when a = 0, then the linker D is attached to the nitrogen atom which carries the radicals R_8 to R_{10},
  - when a = 1, then two of the radicals R_8 to R_{10}, together with the nitrogen atom to which they are attached, form a 5- or 6-membered saturated ring as defined above, and the linker D is carried by a carbon atom of the said saturated ring;

- X^- represents a monovalent or divalent anion and is selected preferably from a halogen atom such as chlorine, bromine, fluorine or iodine, a hydroxide, a hydrogen sulphate or a C_1-C_6 alkyl sulphate such as, for example, a methyl sulphate or an ethyl sulphate;

with the proviso that the number of cationic groups Z is at least 1.

As indicated above, the colorations obtained with the oxidation dyeing composition in accordance with the invention are intense and make it possible to obtain a wide palette of colours. Moreover, they exhibit excellent properties of resistance with respect to the action of various external agents (light, inclement weather, washing, permanent-waving, perspiration, friction). These properties are particularly remarkable as regards notably, the resistance of the colorations obtained with respect to the action of light, washing, permanent-waving and perspiration.

In the formula (I) above, the alkyl and alkoxy radicals may be linear or branched.

Among the rings of the unsaturated groups Z of formula (II) above, mention may be made in particular, by way of example, of pyrrole, imidazole, pyrazole, oxazole, thiazole and triazole rings.

Among the rings of the unsaturated groups Z of formula (III) above, mention may be made in

particular, by way of example, of pyridine, pyrimidine, pyrazine, oxazine and triazine rings.

Among the compounds of formula (I) above, mention may be made in particular of the following:

- 3-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-(2-hydroxyethyl)-3H-imidazol-1-ium chloride,
- 3-[(3-aminopyrazolo[1,5-a]pyrimidin-7-ylcarbamoyl)-methyl]-1-methyl-3H-imidazol-1-ium chloride,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-methylpyridinium methyl sulphate,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-(2-hydroxyethyl)pyridinium chloride,
- 2-[(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)methyl]-1,3-dimethyl-3H-imidazol-1-ium methyl sulphate,
- 3-[(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)-methyl]-1-methylpyridinium methyl sulphate,
- 3-[(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)-methyl]-1-methylpyridinium methyl sulphate,
- 2-(3,7-diamino-5-methylpyrazolo[1,5-a]pyrimidin-6-ylmethyl)-1,3-dimethyl-3H-imidazol-1-ium methyl sulphate,
- 2-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1,3-dimethyl-3H-imidazol-1-ium methyl sulphate,

- 2-(3,7-diaminopyrazolo[1,5-a]pyrimidin-2-yl)-1-methylpyridinium methyl sulphate,
- [3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]trimethylammonium chloride,
- [3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]trimethylammonium methyl sulphate,
- 1-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-methylpiperidinium chloride,
- 1-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-methylpiperidinium methyl sulphate,
- 4-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-4-methylmorpholin-4-ium chloride,
- 4-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-4-methylmorpholin-4-ium methyl sulphate,

and their addition salts with an acid.

Among these compounds of formula (I), more particular preference is given to the following:

- 3-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-(2-hydroxyethyl)-3H-imidazol-1-ium chloride,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-methylpyridinium methyl sulphate,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-(2-hydroxyethyl)pyridinium chloride,

- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-methylpyridinium chloride,
- 4-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-4-methylmorpholin-4-ium chloride,
- 4-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-4-methylmorpholin-4-ium methyl sulphate,

and their addition salts with an acid.

The compounds of formula (I) in accordance with the invention may be readily obtained in accordance with well-known methods of the prior art:

- firstly by reduction of the corresponding cationic nitroso or nitro compounds. In this case, reduction to the corresponding primary aromatic amine is carried out in accordance with conventional methods (J. Lehmman in Houben-Weyl, "Methoden der Organischen Chemie", volume IV/1c: Reduction 1 page 491 to 537, 1980). The methods which are preferred in accordance with the invention involve metals such as Zn, Sn or Fe in an acidic medium such as aqueous hydrochloric acid or aqueous acetic acid in the presence or absence of a cosolvent such as methanol, ethanol or tetrahydrofuran. Catalytic hydrogenation is a preferred reduction method in accordance with the invention. This catalytic hydrogenation makes use of metals such as palladium, platinum or nickel. More particularly still, preference is given to palladium on carbon or to Raney nickel, or else to oxides such as PtO_2 in solvents such as

methanol, ethanol, tetrahydrofuran or ethyl acetate in the presence or absence of an acid such as, for example, acetic acid. These catalytic reductions may also be carried out with formic acid in the presence of a trialkylamine such as triethylamine or with ammonium formate in the place of gaseous hydrogen. (S. Ram, R.E. Ehrenkaufer, Synthesis, 1988,91);

- secondly, by reduction of the corresponding cationic azo compounds (reductive cleavage). Reduction to the corresponding primary aromatic amine is carried out in accordance with conventional methods (J. Lehmman in Houben-Weyl, "Methoden der Organischen Chemie", volume IV/1c: Reduction 1 page 551 to 553, 1980; E.C. Taylor et al., J. Amer. Chem. Soc. 80, 421, 1958).

This step of reduction (to obtain a primary aromatic amine), which gives the synthesized compound its character as an oxidizable compound (oxidation base), and is followed or not by formation of a salt, is in general, for convenience, the last step of the synthesis.

This reduction may occur earlier on in the sequence of reactions leading to the preparation of the compounds of formula (I), and in accordance with well-known processes it is then necessary to "protect" the primary amine created (for example by a step of acetylation, benzenesulphonation, etc.), to carry out subsequently the desired substitution(s) or modification(s) (including the quaternization), and to

end with the "deprotection" (generally in an acidic medium) of the amine function.

When the synthesis is at an end, the compounds of formula (I) in accordance with the invention may, if appropriate, be recovered by well-known methods of the prior art, such as crystallization or distillation.

The invention additionally provides for the use of the compounds of formula (I) in accordance with the invention as oxidation base for the oxidation dyeing of keratinous fibres and, in particular, of human keratinous fibres such as the hair.

The invention also provides a composition for the oxidation dyeing of keratinous fibres and, in particular, of human keratinous fibres such as the hair, characterized in that it comprises as oxidation base, in a medium appropriate for dyeing, at least one compound of formula (I) in accordance with the invention.

The compound or compounds of formula (I) in accordance with the invention represent(s) preferably from 0.0005 to 12% by weight, approximately, of the total weight of the dyeing composition, and, more preferably still, from 0.005 to 6% by weight, approximately, of this weight.

The medium appropriate for dyeing (or vehicle) generally consists of water or a mixture of water and at least one organic solvent for solubilizing

the compounds which would not be sufficiently soluble in water. By way of organic solvent, mention may be made, for example, of C_1-C_4 lower alkanols, such as ethanol and isopropanol; glycerol; glycols and glycol ethers, such as 2-butoxyethanol, propylene glycol, propylene glycol monomethyl ether, diethylene glycol monoethyl and monomethyl ether, and also aromatic alcohols such as benzyl alcohol or phenoxyethanol, similar products and mixtures thereof.

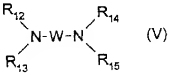
The solvents may be present in proportions of preferably between 1 and 40% by weight, approximately, relative to the total weight of the dyeing composition, and still more preferably between 5 and 30% by weight, approximately.

The pH of the dyeing composition according to the invention is generally between 3 and 12, approximately, and preferably between 5 and 11, approximately. It may be adjusted to the desired value by means of acidifying or basifying agents which are commonly employed in the dyeing of keratinous fibres.

Among the acidifying agents, mention may be made, by way of example, of mineral acids or organic acids such as hydrochloric acid, orthophosphoric acid, sulphuric acid, carboxylic acids such as acetic acid, tartaric acid, citric acid and lactic acid, and sulphonic acids.

Among the basifying agents, mention may be made, by way of example, of aqueous ammonia, alkali

metal carbonates, alkanolamines such as mono-, di- and triethanolamines and their derivatives, sodium hydroxide or potassium hydroxide, and the compounds of formula (V) below:



(V)

in which W is a propylene residue optionally substituted by a hydroxyl group or a C_1-C_6 alkyl radical; and R_{12}, R_{13}, R_{14} and R_{15}, which are identical or different, represent a hydrogen atom or a C_1-C_6 alkyl or C_1-C_6 hydroxyalkyl radical.

Furthermore, the dyeing composition according to the invention may comprise, in addition to the dyes defined above, at least one additional oxidation base, which may be selected from the oxidation bases conventionally used in oxidation dyeing and among which mention may be made in particular of para-phenylenediamines, bisphenylalkylenediamines, para-aminophenols, ortho-aminophenols and heterocyclic bases other than the compounds of formula (I).

Among the para-phenylenediamines, mention may be made more particularly, by way of example, of para-phenylenediamine, para-tolylenediamine, 2,6-dimethyl-para-phenylenediamine, 2-β-hydroxyethyl-para-phenylenediamine, 2-n-propyl-para-phenylenediamine,

2-isopropyl-para-phenylenediamine, N-(β-hydroxypropyl)-para-phenylenediamine, N,N-bis(β-hydroxyethyl)-para-phenylenediamine, 4-amino-N-(β-methoxyethyl)aniline, the para-phenylenediamines described in French Patent Application FR 2 630 438, and their addition salts with an acid.

Among the bisphenylalkylenediamines, mention may be made more particularly, by way of example, of N,N'-bis(β-hydroxyethyl)-N,N'-bis(4'-aminophenyl)-1,3-diaminopropanol, N,N'-bis(β-hydroxyethyl)-N,N'-bis(4'-aminophenyl)ethylenediamine, N,N'-bis(4-aminophenyl)-tetramethylenediamine, N,N'-bis(β-hydroxyethyl)-N,N'-bis(4-aminophenyl)tetramethylenediamine, N,N'-bis(4-methylaminophenyl)tetramethylenediamine, N,N'-bis(ethyl)-N,N'-bis(4'-amino-3'-methylphenyl)-ethylenediamine, and their addition salts with an acid.

Among the para-aminophenols, mention may be made more particularly, by way of example, of para-aminophenol, 4-amino-3-methylphenol, 4-amino-3-fluorophenol, 4-amino-3-hydroxymethylphenol, 4-amino-2-methylphenol, 4-amino-2-hydroxymethylphenol, 4-amino-2-methoxymethylphenol, 4-amino-2-aminomethylphenol, 4-amino-2-(β-hydroxyethylaminomethyl)phenol, and their addition salts with an acid.

Among the ortho-aminophenols, mention may be made more particularly, by way of example, of 2-aminophenol, 2-amino-5-methylphenol, 2-amino-6-

methylphenol, 5-acetamido-2-aminophenol, and their addition salts with an acid.

Among the heterocyclic bases, mention may be made more particularly, by way of example, of the pyridine derivatives, the non-cationic pyrimidine derivatives and the pyrazole derivatives.

Among the pyridine derivatives, mention may be made more particularly of the compounds described, for example, in Patents GB 1 026 978 and GB 1 153 196, such as 2,5-diaminopyridine, 2-(4-methoxyphenyl)amino-3-aminopyridine, 2,3-diamino-6-methoxypyridine, 2-(β-methoxyethyl)amino-3-amino-6-methoxypyridine, 3,4-diaminopyridine, and their addition salts with an acid.

Among the pyrimidine derivatives, mention may be made more particularly of the compounds described, for example, in German Patent DE 2 359 399 or Japanese Patents JP 88-169 571 and JP 91-10659 or Patent Application WO 96/15765, such as 2,4,5,6-tetraamino-pyrimidine, 4-hydroxy-2,5,6-triaminopyrimidine, 2-hydroxy-4,5,6-triaminopyrimidine, 2,4-dihydroxy-5,6-diaminopyrimidine, 2,5,6-triaminopyrimidine, and the pyrazolo-pyrimidine derivatives such as those mentioned in Patent Application FR-A-2 750 048 and among which mention may be made of pyrazolo[1,5-a]pyrimidine-3,7-diamine; 2,5-dimethylpyrazolo[1,5-a]pyrimidine-3,7-diamine; pyrazolo[1,5-a]pyrimidine-3,5-diamine; 2,7-dimethylpyrazolo[1,5-a]pyrimidine-3,5-diamine; 3-aminopyrazolo[1,5-a]pyrimidin-7-ol; 3-aminopyrazolo-

[1,5-a]pyrimidin-5-ol; 2-(3-aminopyrazolo[1,5-a]-pyrimidin-7-ylamino)ethanol, 2-(7-aminopyrazolo[1,5-a]-pyrimidin-3-ylamino)ethanol, 2-[(3-aminopyrazolo-[1,5-a]pyrimidin-7-yl)(2-hydroxyethyl)amino]ethanol, 2-[(7-aminopyrazolo[1,5-a]pyrimidin-3-yl)(2-hydroxy-ethyl)amino]ethanol, 5,6-dimethylpyrazolo-[1,5-a]pyrimidine-3,7-diamine, 2,6-dimethylpyrazolo-[1,5-a]pyrimidine-3,7-diamine, 2,5,N7,N7-tetramethyl-pyrazolo[1,5-a]pyrimidine-3,7-diamine, 3-amino-5-methyl-7-imidazolylpropylamino pyrazolo-[1,5-a]pyrimidine, their addition salts with an acid and their tautomeric forms, when a tautomeric equilibrium exists.

Among the pyrazole derivatives, mention may be made more particularly of the compounds described in Patents DE 3 843 892, DE 4 133 957 and Patent Applications WO 94/08969, WO 94/08970, FR-A-2 733 749 and DE 195 43 988, such as 4,5-diamino-1-methyl-pyrazole, 3,4-diaminopyrazole, 4,5-diamino-1-(4'-chlorobenzyl)pyrazole, 4,5-diamino-1,3-dimethyl-pyrazole, 4,5-diamino-3-methyl-1-phenylpyrazole, 4,5-diamino-1-methyl-3-phenylpyrazole, 4-amino-1,3-dimethyl-5-hydrazinopyrazole, 1-benzyl-4,5-diamino-3-methylpyrazole, 4,5-diamino-3-tert-butyl-1-methylpyrazole, 4,5-diamino-1-tert-butyl-3-methylpyrazole, 4,5-diamino-1-(β-hydroxyethyl)-3-methylpyrazole, 4,5-diamino-1-ethyl-3-methylpyrazole, 4,5-diamino-1-ethyl-3-(4'-methoxyphenyl)pyrazole,

4,5-diamino-1-ethyl-3-hydroxymethylpyrazole, 4,5-diamino-3-hydroxymethyl-1-methylpyrazole, 4,5-diamino-3-hydroxymethyl-1-isopropylpyrazole, 4,5-diamino-3-methyl-1-isopropylpyrazole, 4-amino-5-(2'-aminoethyl)amino-1,3-dimethylpyrazole, 3,4,5-triaminopyrazole, 1-methyl-3,4,5-triaminopyrazole, 3,5-diamino-1-methyl-4-methylaminopyrazole, 3,5-diamino-4-(β-hydroxyethyl)amino-1-methylpyrazole, and their addition salts with an acid.

When they are used, these additional oxidation bases represent preferably from 0.0005 to 12% by weight, approximately, of the total weight of the dyeing composition, and more preferably still from 0.005 to 6% by weight, approximately, of this weight.

The oxidation dyeing compositions according to the invention may also include at least one coupler and/or at least one direct dye, in particular for the purpose of modifying the shades or enriching them with glints.

The couplers which may be used in the oxidation dyeing compositions according to the invention may be selected from the couplers used conventionally in oxidation dyeing, among which mention may be made in particular of meta-phenylenediamines, meta-aminophenols, meta-diphenols and the heterocyclic couplers such as, for example, the indole derivatives, indoline derivatives, pyridine derivatives and pyrazolones, and their addition salts with an acid.

These couplers are selected more particularly from 2-methyl-5-aminophenol, 5-N-(β-hydroxyethyl)amino-2-methylphenol, 3-aminophenol, 1,3-dihydroxybenzene, 1,3-dihydroxy-2-methylbenzene, 4-chloro-1,3-dihydroxybenzene, 2,4-diamino-1-(β-hydroxyethyloxy)-benzene, 2-amino-4-(β-hydroxyethylamino)-1-methoxy-benzene, 1,3-diaminobenzene, 1,3-bis(2,4-diamino-phenoxy)propane, sesamol, α-naphthol, 6-hydroxyindole, 4-hydroxyindole, 4-hydroxy-N-methylindole, 6-hydroxy-indoline, 2,6-dihydroxy-4-methylpyridine, 1H-3-methylpyrazol-5-one, 1-phenyl-3-methylpyrazol-5-one, and their addition salts with an acid.

When they are present, these couplers represent preferably from 0.0001 to 10% by weight, approximately, of the total weight of the dyeing composition, and more preferably still from 0.005 to 5% by weight, approximately, of this weight.

Generally speaking, the addition salts with an acid which may be used in the context of the invention (compounds of formula (I), additional oxidation bases and couplers) are selected in particular from the hydrochlorides, hydrobromides, sulphates, citrates, succinates, tartrates, lactates and acetates.

The dyeing composition in accordance with the invention may also include various adjuvants which are conventionally employed in hair-dyeing compositions, such as anionic, cationic, nonionic, amphoteric or

zwitterionic surfactants or mixtures thereof, anionic, cationic, nonionic, amphoteric or zwitterionic polymers or mixtures thereof, mineral or organic thickeners, antioxidants, penetrants, sequestrants, perfumes, buffers, dispersants, conditioning agents such as, for example, silicones, film-formers, preservatives and opacifiers.

The person skilled in the art will of course take care to select this or these optional complementary compounds such that the advantageous properties intrinsically associated with the oxidation dyeing composition in accordance with the invention are not, or not substantially, adversely affected by the intended addition or additions.

The dyeing composition according to the invention may be presented in a variety of forms, such as in the form of liquids, creams, gels, or in any other form appropriate for carrying out dyeing of keratinous fibres and, in particular, of human hair.

The invention additionally provides a method of dyeing keratinous fibres and, in particular, human keratinous fibres such as the hair, employing the dyeing composition as defined above.

In accordance with this method, at least one dyeing composition as defined above is applied to the fibres, the colour being revealed at an acidic, neutral or alkaline pH with the aid of an oxidizing agent which is added to the dyeing composition right at the time of

use or which is present in an oxidizing composition which is applied simultaneously or sequentially, separately.

In accordance with a preferred embodiment of the dyeing method of the invention, the dyeing composition described above is preferably mixed at the time of use with an oxidizing composition comprising, in a medium appropriate for dyeing, at least one oxidizing agent present in an amount sufficient to develop a coloration. The mixture obtained is then applied to the keratinous fibres and is left to act for from approximately 3 to 50 minutes, preferably for from approximately 5 to 30 minutes, after which the fibres are rinsed, washed with shampoo, rinsed again and dried.

The oxidizing agent may be selected from the oxidizing agents which are conventionally used for the oxidation dyeing of keratinous fibres, among which mention may be made of hydrogen peroxide, urea peroxide, alkali metal bromates, persalts such as perborates and persulphates, and enzymes such as peroxidases, laccases, tyrosinases and oxidoreductases, among which mention may be made in particular of the pyranose oxidases, glucose oxidases, glycerol oxidases, lactate oxidases, pyruvate oxidases and uricases.

The pH of the oxidizing composition comprising the oxidizing agent as defined above is such that, after mixing with the dyeing composition, the pH

of the resultant composition that is applied to keratinous fibres varies preferably between approximately 3 and 12, and more preferably still between 5 and 11. It is adjusted to the desired value by means of acidifying or basifying agents which are commonly employed in dyeing keratinous fibres, such agents being as defined above.

The oxidizing composition as defined above may also include various adjuvants which are conventionally used in hair-dyeing compositions, such adjuvants being as defined above.

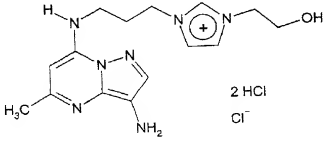
The composition which is ultimately applied to the keratinous fibres may be presented in various forms, such as in the form of liquids, creams or gels or in any other form appropriate for carrying out dyeing of keratinous fibres and, in particular, of human hair.

The invention also provides a multi-compartment dyeing device or "kit", or any other packaging system having two or more compartments, of which a first compartment contains the dyeing composition as defined above and a second compartment contains the oxidizing composition as defined above. These devices may be equipped with a means allowing the desired mixture to be delivered to the hair, such as the devices described in Patent FR-2 586 913 in the name of the applicant.

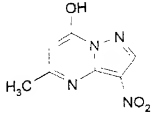
The examples which follow are intended to illustrate the invention without limiting its scope.

PREPARATION EXAMPLES

PREPARATION EXAMPLE 1: Synthesis of 3-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-(2-hydroxyethyl)-3H-imidazol-1-ium dihydrochloride



A) Preparation of 3-nitro-5-methylpyrazolo[1,5-a]pyrimidin-7-ol

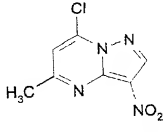


50 g of 4-nitro-2H-pyrazol-3-ylamine hydrochloride (prepared in accordance with H. Dorn and H. Dilcher, Liebigs Ann. Chem., 707, 141, 1967) and 60 g of ethyl acetoacetate in 160 cc of acetic acid were introduced into a 500 cc three-necked round-bottomed flask equipped with a magnetic stirrer, a thermometer and a condenser. The reaction medium was refluxed for 12 hours. The precipitate which formed was filtered off at about 90°C. It was washed with diisopropyl ether and dried under vacuum over

phosphoric anhydride. This gave 50 g of 3-nitro-5-methylpyrazolo[1,5-a]pyrimidin-7-ol in the form of yellow crystals (yield = 84.5%; melting point = 290°C with decomposition), whose elemental analysis, calculated for $C_7H_6N_4O_3$ (MW = 194.15 g), was as follows:

%	C	H	N	O
Calculated	43.31	3.12	28.86	24.72
Found	43.12	3.11	28.77	24.65

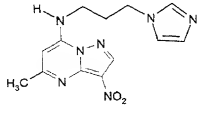
b) Preparation of 7-chloro-5-methyl-3-nitropyrazolo-[1,5-a]pyrimidine



230 cc of phosphorus oxychloride, 15.4 g of N,N-dimethylaniline and 23.3 g of 3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ol were introduced into a 500 cc three-necked flask equipped with a magnetic stirrer, a thermometer and a condenser. The reaction medium was refluxed for 2.5 h. Following evaporation of the phosphorus oxychloride under reduced pressure, a highly viscous green oil was obtained to which approximately 400 g of ice were added. A brown solid precipitated. After 30 minutes of stirring, the precipitate was filtered off and washed with petroleum

ether and then with diisopropyl ether. Drying under vaccum over phosphoric anhydride gave 21.4 g of 7-chloro-5-methyl-3-nitropyrazolo[1,5-a]pyrimidine in the form of a brown solid with a yield of 83.9%.

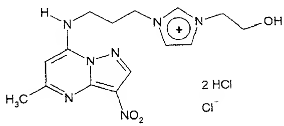
c) Preparation of (3-imidazol-1-ylpropyl)(5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-7-yl)amine



2.88 g of 3-imidazol-1-ylpropylamine and 2.33 g of triethylamine in 20 cc of dioxane were introduced into a 100 cc three-necked round-bottomed flask equipped with a magnetic stirrer, a thermometer, a dropping funnel and a condenser. 4.5 g of 7-chloro-5-methyl-3-nitropyrazolo[1,5-a]pyrimidine in solution in 20 cc of dioxane and 5 cc of dimethylformamide were added dropwise. After 2 hours of stirring at room temperature, the precipitate was filtered off. It was washed with diisopropyl ether and dried under vacuum. This gave 7.2 g of crude product. This product was taken up under reflux in 28 cc of water, then filtered off at room temperature. This operation was repeated a second time. The product was washed with ethanol and with diisopropyl ether. This gave 4.1 g of (3-imidazol-1-ylpropyl)(5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-7-

yl)amine in the form of a beige powder, after drying under vacuum over phosphoric anhydride, with a yield of 65%.

d) <u>Preparation of 1-(2-hydroxyethyl)-3-[3-(5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-3H-imidazol-1-ium chloride</u>



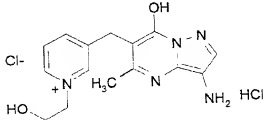
3 g of (3-imidazol-1-ylpropyl)(5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-7-yl)amine and 10 g of 2-chloroethanol were introduced into a 25 cc three-necked round-bottomed flask equipped with a magnetic stirrer, a thermometer and a condenser. The medium was refluxed for 6 hours. The reaction medium was poured into 160 cc of ethyl acetate and refluxed. The precipitate was filtered off at room temperature. This gave 3.8 g of 1-(2-hydroxyethyl)-3-[3-(5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-3H-imidazol-1-ium chloride (beige powder) after drying under vacuum over phosphoric anhydride.

e) Preparation of 3-[3-(3-amino-5-methylpyrazolo-[1,5-a]pyrimidin-7-ylamino)propyl]-1-(2-hydroxyethyl)-3H-imidazol-1-ium chloride dihydrochloride

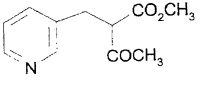
3.5 g of 1-(2-hydroxyethyl)-3-[3-(5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-3H-imidazol-1-ium chloride in 150 cc of ethanol and then 0.39 g of 5% palladium on carbon (containing 50% water) were introduced into a 250 cc hydrogenator. Between 11 and 12 bars' pressure of hydrogen were introduced into the reactor and the reaction medium was brought to 60°C. After 4 hours of reaction, the catalyst was filtered over Celite and a stream of gaseous hydrochloric acid was passed through the filtrate. The reaction medium was poured into 100 cc of diisopropyl ether. After stirring, the precipitate was filtered off. It was washed with diisopropyl ether and dried under vacuum over phosphoric anhydride. This gave 3.3 g of a highly hygroscopic product. A 3% aqueous solution of this product was formed, and was lyophilized. The resulting solid was taken up at reflux in 30 cc of absolute ethanol. This gives 2.25 g of 3-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimdin-7-ylamino)propyl]-1-(2-hydroxyethyl)-3H-imidazol-1-ium chloride in the form of the dihydrochloride, after drying under vacuum over phosphoric anhydride, with a yield of 85%, whose analysis calculated for $C_{15}H_{22}N_7OCl$, 2HCl (MW = 424.76 g) was as follows:

%	C	H	N	O	Cl
Calculated	42.42	5.70	23.08	3.77	25.04
Found	40.28	6.19	21.40	7.99	24.14
Calculated with 1 mol of water	40.69	5.92	22.14	7.23	24.02

PREPARATION EXAMPLE 2: Synthesis of 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]pyrimidin-6-ylmethyl)-1-(2-hydroxyethyl)pyridinium chloride, hydrochloride



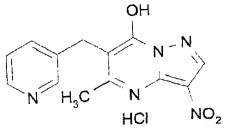
a) Preparation of the methyl ester of 3-oxo-2-pyridin-3-ylmethylbutyric acid



25 g of the methyl ester of 2-acetyl-3-pyridin-3-ylacrylic acid (prepared in accordance with I. Adachi et al., Chem. Pharm. Bull. 35(8), 3235, 1987), 200 cc of ethanol and 5.25 g of 5% palladium on carbon (containing 50% water) were introduced into a 300 cc hydrogenation reactor. A hydrogen pressure of 6 bars was introduced and the reduction was carried out at ambient temperature. The reaction medium was treated

when there was no longer any absorption of hydrogen. The catalyst was filtered off and the solvent was evaporated. This gave 24 g of crude product which was treated with 200 cc of diethyl ether. The white precipitate was filtered off and the solvent was evaporated. This gave 20 g of the methyl ester of 3-oxo-2-pyridin-3-ylmethylbutyric acid in the form of a brown oil, with a yield of 79%.

b) Preparation of 5-methyl-3-nitro-6-pyridin-3-yl-methylpyrazolo[1,5-a]pyrimidin-7-ol hydrochloride

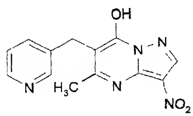


89 g of 4-nitro-2H-pyrazol-3-ylamine (prepared in accordance with H. Dorm and H. Dilcher, Liebigs Ann. Chem., 707, 141, 1967) and 112 g of the methyl ester of 3-oxo-2-pyridin-3-ylmethylbutyric acid, obtained above in the preceding step, in 1120 cc of acetic acid were introduced into a 2-litre three-necked round-bottomed flask equipped with a magnetic stirrer, a thermometer and a condenser. The reaction medium was refluxed for 5 hours. The precipitate which formed was filtered off at room temperature. It was rinsed with diisopropyl ether and dried under vacuum over

phosphoric anhydride. This gave 120 g of crude product. This was recrystallized from a water/acetone (1/25) mixture. This gave 77 g of 5-methyl-3-nitro-6-pyridin-3-ylmethylpyrazolo[1,5-a]pyrimidin-7-ol hydrochloride in the form of yellow crystals (yield = 50%), whose elemental analysis calculated for $C_{13}H_{11}N_5O_3$.HCl was as follows:

%	C	H	N	O	Cl
Calculated	48.53	3.76	21.77	14.92	11.02
Found	48.31	3.82	21.89	14.23	11.75

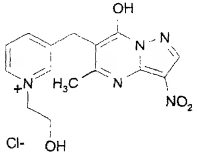
c) Preparation of 5-methyl-3-nitro-6-pyridin-3-ylmethylpyrazolo[1,5-a]pyrimidin-7-ol



250 cc of water and 10.6 g of 20% aqueous ammonia were introduced into a 500-cc Erlenmeyer flask equipped with a magnetic stirrer. 20 g of 5-methyl-3-nitro-6-pyridin-3-ylmethylpyrazolo[1,5-a]pyrimidin-7-ol hydrochloride were added in solid portions. The reaction mixture was left with stirring at room temperature for 3 hours. The resulting solid was filtered off and then washed with 100 cc of water and then with diisopropyl ether. The product was dried over phosphoric anhydride. This gave 16 g of 5-methyl-3-

nitro-6-pyridin-3-ylmethylpyrazolo[1,5-a]pyrimidin-7-ol with a yield of 90%.

d) Preparation of 1-(2-hydroxyethyl)-3-(7-hydroxy-5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-6-ylmethyl)-pyridinium chloride



10 g of 5-methyl-3-nitro-6-pyridin-3-yl-methylpyrazolo[1,5-a]pyrimidin-7-ol, obtained above in the preceding step, and 100 cc of 2-chloroethanol were introduced into a 250-cc three-necked round-bottomed flask equipped with a magnetic stirrer, a thermometer and a condenser. The medium was refluxed for 5 hours. The solvent was evaporated and then the product was treated with ethanol. The precipitate was filtered off at room temperature. This gave 10 g of crude product. It was recrystallized from acetic acid. This gave 6.7 g of 1-(2-hydroxyethyl)-3-(7-hydroxy-5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-6-ylmethyl)pyridinium chloride after drying under vacuum over phosphoric anhydride (yield = 52%), whose elemental analysis calculated for $C_{15}H_{16}N_5O_4.Cl$ with 0.28 mol of acetic acid was as follows:

%	C	H	N	O	Cl
Calculated	48.80	4.47	18.29	19.07	9.28
Found	47.69	4.56	18.26	18.85	9.51

e) Preparation of 3-(3-amino-7-hydroxy-5-methyl-pyrazolo[1,5-a]pyrimidin-6-ylmethyl)-1-(2-hydroxy-ethyl)pyridinium chloride, hydrochloride

2 g of 1-(2-hydroxyethyl)-3-(7-hydroxy-5-methyl-3-nitropyrazolo[1,5-a]pyrimidin-6-ylmethyl)-pyridinium chloride obtained above in the preceding step in 200 cc of acetic acid and then 0.6 g of 5% palladium on carbon (containing 50% water) were introduced into a 500 cc hydrogenator. 8 bars of hydrogen pressure were introduced into the reactor and the reaction medium was brought to 50°C. After reaction for 3 hours, the catalyst was filtered off over Celite. The solvent was evaporated and the crude product obtained was taken up in 10 cc of 7M hydrochloric ethanol. The precipitate was filtered off. It was washed with diisopropyl ether and dried under vacuum over phosphoric anhydride. This gave 2.7 g of 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]pyrimidin-6-yl-methyl)-1-(2-hydroxyethyl)pyridinium chloride (a highly hygroscopic product) in the form of the hydrochloride, after drying under vacuum over phosphoric anhydride, with a yield of 75% and an analysis, calculated for $C_{15}H_{18}N_5O_2Cl$. HCl, which was as follows:

%	C	H	N	O	Cl
Calculated	48.40	5.14	18.81	8.60	19.05
Found	47.04	5.25	17.59	10.26	18.40
Calculated with 0.5 mol of water	47.20	5.24	18.34	10.48	18.61

APPLICATION EXAMPLES

EXAMPLES 1 TO 7 OF DYEING IN A BASIC MEDIUM

The following dyeing compositions (amounts in grams) were prepared:

EXAMPLE	1	2	3	4	5	6	7
3-[3-(3-Amino-5-methylpyrazolo[1,5-a]-pyrimidin-7-ylamino)propyl]-1-(2-hydroxy-ethyl)-3H-imidazol-1-ium chloride dihydro-chloride (oxidation base of formula (I))	1.27	1.27	1.27	1.27	1.27	1.27	1.27
1,3-Dihydroxybenzene (coupler)	-	0.33	-	-	-	-	-
3-Aminophenol (coupler)	-	-	0.327	-	-	-	-
5-N-(β-Hydroxyethyl)amino 2-methylphenol (coupler)	-	-	-	0.504	-	-	-
2,4-Diamino 1-(β-hydroxyethyloxy)benzene, 2HCl (coupler)	-	-	-	-	0.723	-	-
6-Hydroxyindole (coupler)	-	-	-	-	-	0.399	-
4-Hydroxyindole (coupler)	-	-	-	-	-	-	0.399
Common dyeing vehicle 1	(*)	(*)	(*)	(*)	(*)	(*)	(*)
Demineralized water q.s.	100 g	100 g	100 g	100 g	100 g	100 g	100 g

(*) Common dyeing vehicle 1:

- 96° ethyl alcohol 18 g
- Sodium metabisulphite in 35% aqueous solution 0.68 g
- Pentasodium salt of diethylenetriaminepentaacetic acid 1.1 g
- 20% aqueous ammonia 10.0 g

Each of the above dyeing compositions was mixed at the time of use, weight for weight, with a 20-volume hydrogen peroxide solution (6% by weight) with a pH of 3.

The mixture obtained was applied to locks of permed grey hair containing 90% white hair, for 30 minutes. The locks were subsequently rinsed, washed with a standard shampoo, rinsed again and then dried.

The shades obtained are given in the table below:

EXAMPLE	Dyeing pH	Shade obtained
1	10 ± 0.2	iridescent dark blonde
2	10 ± 0.2	coppery red
3	10 ± 0.2	purple mahogany
4	10 ± 0.2	red copper
5	10 ± 0.2	deep violet irridescent light chestnut
6	10 ± 0.2	natural chestnut
7	10 ± 0.2	deep red irridescent

EXAMPLES 8 TO 14 OF DYEING IN A NEUTRAL MEDIUM

The following dyeing compositions (amounts in grams) were prepared:

EXAMPLE	8	9	10	11	12	13	14
3-[3-(3-Amino-5-methylpyrazolo[1,5-a]-pyrimidin-7-ylamino)propyl]-1-(2-hydroxy-ethyl)-3H-imidazol-1-ium chloride dihydro-chloride (oxidation base of formula (I))	1.27	1.27	1.27	1.27	1.27	1.27	1.27
1,3-Dihydroxybenzene (coupler)	-	0.33	-	-	-	-	-
3-Aminophenol (coupler)	-	-	0.327	-	-	-	-
5-N-(β-Hydroxyethyl)amino 2-methylphenol (coupler)	-	-	-	0.504	-	-	-
2,4-Diamino 1-(β-hydroxyethyloxy)benzene, 2HCl (coupler)	-	-	-	-	0.723	-	-
6-Hydroxyindole (coupler)	-	-	-	-	-	0.399	-
4-Hydroxyindole (coupler)	-	-	-	-	-	-	0.399
Common dyeing vehicle 2	(**)	(**)	(**)	(**)	(**)	(**)	(**)
Demineralized water q.s.	100 g	100 g	100 g	100 g	100 g	100 g	100 g

(**) Common dyeing vehicle 2:

- 96° ethanol 18 g
- K_2HPO_4/KH_2PO_4 (1.5M/1M) buffer
- Sodium metabisulphite 10 g
- Pentasodium salt of diethylenetriaminepentaacetic acid 0.68 g

Each of the above dyeing compositions was mixed at the time of use, weight for weight, with a 20-volume hydrogen peroxide solution (6% by weight) with a pH of 3.

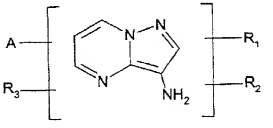
The mixture obtained was applied to locks of natural grey hair containing 90% white hair, for 30 minutes. The locks were subsequently rinsed, washed with a standard shampoo, rinsed again and then dried.

The shades obtained are given in the table below:

EXAMPLE	Dyeing pH	Shade obtained
8	5.7 ± 0.2	coppery iridescent blonde
9	5.7 ± 0.2	beige light ash blonde
10	5.7 ± 0.2	coppery brown dark blonde
11	5.7 ± 0.2	coppery dark blonde
12	5.7 ± 0.2	purplish red
13	5.7 ± 0.2	natural dark ash blonde
14	5.7 ± 0.2	reddish purple

CLAIMS

1. Compounds of formula (I) below, and their addition salts with an acid:



(I)

in which:

- R_1, R_2 and R_3, which may be identical or different, represent a hydrogen atom; a halogen atom; a group Z; a (C_1-C_6 alkyl)carbonyl radical; an amino(C_1-C_6 alkyl)carbonyl radical; an N-Z-amino(C_1-C_6 alkyl)carbonyl radical; an N-(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl radical; an N,N-di(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl radical; an amino(C_1-C_6 alkyl)carbonyl(C_1-C_6 alkyl) radical; an N-Z-amino(C_1-C_6 alkyl)carbonyl(C_1-C_6 alkyl) radical; an N-(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl(C_1-C_6 alkyl) radical; an N,N-di(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl(C_1-C_6 alkyl) radical; a carboxyl radical; a (C_1-C_6 alkyl)carboxyl radical; a (C_1-C_6 alkyl)sulphonyl radical; an aminosulphonyl radical; an N-Z-aminosulphonyl radical; an N-(C_1-C_6 alkyl)aminosulphonyl radical; an N,N-di(C_1-C_6 alkyl)-aminosulphonyl radical; an aminosulphonyl(C_1-C_6 alkyl) radical; an N-Z-aminosulphonyl(C_1-C_6 alkyl) radical; an N-(C_1-C_6 alkyl)aminosulphonyl(C_1-C_6 alkyl) radical;

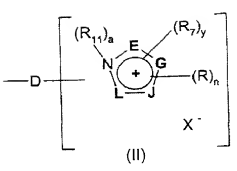
an N,N-di(C_1-C_6 alkyl)aminosulphonyl(C_1-C_6 alkyl) radical; a carbamyl radical; an N-(C_1-C_6 alkyl)carbamyl radical; an N,N-di(C_1-C_6 alkyl)carbamyl radical; a carbamyl(C_1-C_6 alkyl) radical; an N-(C_1-C_6 alkyl)carbamyl(C_1-C_6 alkyl) radical; an N,N-di(C_1-C_6 alkyl)carbamyl(C_1-C_6 alkyl) radical; a C_1-C_6 alkyl radical; a hydroxyl radical; a nitro radical; a C_1-C_6 monohydroxyalkyl radical; a C_2-C_6 polyhydroxyalkyl radical; a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical; a C_1-C_6 trifluoroalkyl radical; a cyano radical; a group OR_6 or SR_6; an amino radical; an N-(C_1-C_6 alkyl)amino radical; an N,N-di(C_1-C_6 alkyl)amino radical (where the two alkyl substituents may form a 5- or 6-membered ring); an N-hydroxy(C_1-C_6 alkyl)amino radical; an N,N-bis(hydroxy(C_1-C_6 alkyl))amino radical; an N-polyhydroxy(C_2-C_6 alkyl)amino radical; an N,N-bis(polyhydroxy(C_2-C_6 alkyl))amino radical; an amino(C_1-C_6 alkyl)amino radical in which the terminal amino group is unsubstituted or substituted by one or two C_1-C_6 alkyl radicals, where the said alkyl radicals may form a saturated or unsaturated, 5- or 6-membered ring; an amino group protected by a (C_1-C_6 alkyl)carbonyl, trifluoro(C_1-C_6 alkyl)carbonyl, amino(C_1-C_6 alkyl)carbonyl, N-Z-amino(C_1-C_6 alkyl)-carbonyl, N-(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl, N,N-di(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl or formyl radical or by a group Z;

- R_6 denotes a C_1-C_6 alkyl radical; a C_1-C_6 monohydroxyalkyl radical; a C_2-C_6 polyhydroxyalkyl radical; a group Z; a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical; an aryl radical; a benzyl radical; a C_1-C_6 carboxyalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carboxyalkyl radical; a C_1-C_6 cyanoalkyl radical; a C_1-C_6 carbamylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 N,N-di(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 trifluoroalkyl radical; a C_1-C_6 aminosulphonylalkyl radical; a C_1-C_6 N-Z-aminosulphonylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)aminosulphonylalkyl radical; a C_1-C_6 N,N-di(C_1-C_6 alkyl)aminosulphonylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)sulphinylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)sulphonylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carbonylalkyl radical; a C_1-C_6 aminoalkyl radical; a C_1-C_6 aminoalkyl radical whose amine is substituted by one or two identical or different radicals selected from C_1-C_6 alkyl, monohydroxy(C_1-C_6 alkyl), polyhydroxy(C_2-C_6 alkyl), (C_1-C_6 alkyl)-carbonyl, formyl, trifluoro(C_1-C_6 alkyl)carbonyl and (C_1-C_6 alkyl)sulphonyl radicals or by a group Z;
- A represents a group $-NR_4R_5$ or a hydroxyl radical;
- R_4 and R_5, which are identical or different, represent a hydrogen atom; a group Z; a C_1-C_6 alkyl radical; a C_1-C_6 monohydroxyalkyl radical; a C_2-C_6 polyhydroxyalkyl radical; a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical; an aryl radical; a benzyl radical; a C_1-C_6

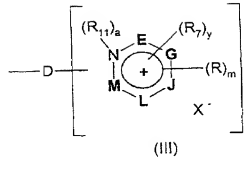
cyanoalkyl radical; a C_1-C_6 carbamylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 N,N-di(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 thiocarbamylalkyl radical; a C_1-C_6 trifluoroalkyl radical; a C_1-C_6 sulphoalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carboxyalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)sulphinylalkyl radical; a C_1-C_6 aminosulphonylalkyl radical; a C_1-C_6 N-Z-aminosulphonylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)-aminosulphonylalkyl radical; a C_1-C_6 N,N-di(C_1-C_6 alkyl)aminosulphonylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carbonylalkyl radical; a C_1-C_6 aminoalkyl radical; a C_1-C_6 aminoalkyl radical whose amine is substituted by one or two identical or different radicals selected from C_1-C_6 alkyl, C_1-C_6 monohydroxyalkyl, C_2-C_6 polyhydroxyalkyl, (C_1-C_6 alkyl)carbonyl, (C_1-C_6 alkyl)sulphonyl, formyl and trifluoro(C_1-C_6 alkyl)carbonyl radicals or by a group Z;

one and only one of the radicals R_4 and R_5 may also represent a (C_1-C_6 alkyl)carbonyl; formyl; trifluoro(C_1-C_6 alkyl)carbonyl; amino(C_1-C_6 alkyl)carbonyl, N-Z-amino(C_1-C_6 alkyl)carbonyl; N-(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl; or N,N-di(C_1-C_6 alkyl)amino(C_1-C_6 alkyl)carbonyl radical;

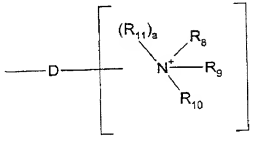
- Z is selected from the unsaturated cationic groups of formulae (II) and (III) below and the saturated cationic groups of formula (IV) below:



(II)



(III)



(IV)

in which:

- D is a linker which represents an alkyl chain containing preferably 1 to 14 carbon atoms, is linear or branched and may be interrupted by one or more heteroatoms such as oxygen, sulphur or nitrogen atoms and may be substituted by one or more hydroxyl or C_1-C_6 alkoxy radicals, and may carry one or more ketone functions;
- the ring members E, G, J, L and M, which are identical or different, represent a carbon, oxygen, sulphur or nitrogen atom;
- n is an integer between 0 and 4, inclusively;
- m is an integer between 0 and 5, inclusively;
- the radicals R, which are identical or different, represent a group Z, a halogen atom, a hydroxyl radical, a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical, a C_2-C_6 polyhydroxyalkyl

radical, a nitro radical, a cyano radical, a C_1-C_6 cyanoalkyl radical, a C_1-C_6 alkoxy radical, a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical, an amido radical, an aldehydo radical, a carboxyl radical, a C_1-C_6 alkylcarbonyl radical, a thio radical, a C_1-C_6 thioalkyl radical, a (C_1-C_6 alkyl)thio radical, an amino radical, an amino radical protected by a (C_1-C_6 alkyl)carbonyl, carbamyl or (C_1-C_6 alkyl)sulphonyl radical; a group NHR" or NR"R"' in which R" and R"', which are identical or different, represent a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical or a C_2-C_6 polyhydroxyalkyl radical;

- R_7 represents a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical, a C_2-C_6 polyhydroxyalkyl radical, a C_1-C_6 cyanoalkyl radical, a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical, a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical, a carbamyl(C_1-C_6 alkyl) radical, a C_1-C_6 (C_1-C_6 alkyl)carboxyalkyl radical, a benzyl radical, or a group Z;
- R_8, R_9 and R_{10}, which are identical or different, represent a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical, a C_2-C_6 polyhydroxyalkyl radical, a C_1-C_6 (C_1-C_6 alkoxy)alkyl radical, a C_1-C_6 cyanoalkyl radical, an aryl radical, a benzyl radical, a C_1-C_6 amidoalkyl radical, a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical or a C_1-C_6 aminoalkyl radical whose amine is protected by a (C_1-C_6 alkyl)carbonyl, amido, carboxyl or (C_1-C_6

alkyl)sulphonyl radical; two of the radicals R_8, R_9 and R_{10} may also form, together with the nitrogen atom to which they are attached, a saturated 5- or 6-membered carbon-containing ring or one containing one or more heteroatoms, it being possible for the said ring to be unsubstituted or substituted by a halogen atom, a hydroxyl radical, a C_1-C_6 alkyl radical, a C_1-C_6 monohydroxyalkyl radical, a C_2-C_6 polyhydroxyalkyl radical, a nitro radical, a cyano radical, a C_1-C_6 cyanoalkyl radical, a C_1-C_6 alkoxy radical, a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical, an amido radical, an aldehydo radical, a carboxyl radical, a C_1-C_6 ketoalkyl radical, a thio radical, a C_1-C_6 thioalkyl radical, a (C_1-C_6 alkyl)thio radical, an amino radical, or an amino radical protected by a (C_1-C_6 alkyl)carbonyl, carbamyl or (C_1-C_6 alkyl)sulphonyl radical;

one of the radicals R_8, R_9 and R_{10} may also represent a second group Z, identical to or different from the first group Z;

- R_{11} represents a C_1-C_6 alkyl radical; a C_1-C_6 monohydroxyalkyl radical; a C_2-C_6 polyhydroxyalkyl radical; an aryl radical; a benzyl radical; a C_1-C_6 aminoalkyl radical, a C_1-C_6 aminoalkyl radical whose amine is protected by a (C_1-C_6 alkyl)carbonyl, carbamyl or (C_1-C_6 alkyl)sulphonyl radical; a C_1-C_6 carboxyalkyl radical; a C_1-C_6 cyanoalkyl radical; a C_1-C_6 carbamylalkyl radical; a C_1-C_6 trifluoroalkyl

radical; a C_1-C_6 tri(C_1-C_6 alkyl)silanealkyl radical; a C_1-C_6 sulphonamidoalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)carboxyalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)-sulphinylalkyl radical; a C_1-C_6 (C_1-C_6 alkyl)sulphonyl-alkyl radical; a C_1-C_6 (C_1-C_6 alkyl)ketoalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)carbamylalkyl radical; a C_1-C_6 N-(C_1-C_6 alkyl)sulphonamidoalkyl radical;

- a and y are integers equal to 0 or 1; with the following conditions:
  - in the unsaturated cationic groups of formula (II):
    - when a = 0, the linker D is attached to the nitrogen atom,
    - when a = 1, the linker D is attached to one of the ring members E, G, J or L,
    - y can adopt the value 1 only
      1) when the ring members E, G, J and L simultaneously represent a carbon atom and when the radical R_7 is carried by the nitrogen atom of the unsaturated ring; or else
      2) when at least one of the ring members E, G, J and L represents a nitrogen atom to which the radical R_7 is attached;
  - in the unsaturated cationic groups of formula (III):
    - when a = 0, the linker D is attached to the nitrogen atom,
    - when a = 1, the linker D is attached to one of the ring members E, G, J, L or M,

- y can adopt the value 1 only when at least one of the ring members E, G, J, L and M represents a divalent atom and when the radical R_7 is carried by the nitrogen atom of the unsaturated ring;

- in the cationic groups of formula (IV):
  - when a = 0, then the linker D is attached to the nitrogen atom which carries the radicals R_8 to R_{10},
  - when a = 1, then two of the radicals R_8 to R_{10}, together with the nitrogen atom to which they are attached, form a 5- or 6-membered saturated ring as defined above, and the linker D is carried by a carbon atom of the said saturated ring;

- X^- represents a monovalent or divalent anion;

with the proviso that the number of cationic groups Z is at least 1.

2. Compounds according to Claim 1, characterized in that the rings of the unsaturated groups Z of formula (II) are selected from pyrrole, imidazole, pyrazole, oxazole, thiazole and triazole rings.

3. Compounds according to Claim 1, characterized in that the rings of the unsaturated groups Z of formula (III) are selected from pyridine, pyrimidine, pyrazine, oxazine and triazine rings.

4. Compounds according to any one of the preceding claims, characterized in that two of the radicals R_8, R_9 and R_{10} form a pyrrolidine ring, a piperidine ring, a piperazine ring or a morpholine ring.

5. Compounds according to any one of the preceding claims, characterized in that X^- represents a halogen atom, a hydroxide, a hydrogen sulphate or a C_1-C_6 alkyl sulphate.

6. Compounds according to any one of the preceding claims, characterized in that they are selected from:

- 3-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-(2-hydroxyethyl)-3H-imidazol-1-ium chloride,
- 3-[(3-aminopyrazolo[1,5-a]pyrimidin-7-ylcarbamoyl)-methyl]-1-methyl-3H-imidazol-1-ium chloride,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-methylpyridinium methyl sulphate,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-(2-hydroxyethyl)pyridinium chloride,
- 2-[(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)methyl]-1,3-dimethyl-3H-imidazol-1-ium methyl sulphate,
- 3-[(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)-methyl]-1-methylpyridinium methyl sulphate,

- 3-[(3-aminopyrazolo[1,5-a]pyrimidin-7-ylamino)-methyl]-1-methylpyridinium methyl sulphate,
- 2-(3,7-diamino-5-methylpyrazolo[1,5-a]pyrimidin-6-ylmethyl)-1,3-dimethyl-3H-imidazol-1-ium methyl sulphate,
- 2-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1,3-dimethyl-3H-imidazol-1-ium methyl sulphate,
- 2-(3,7-diaminopyrazolo[1,5-a]pyrimidin-2-yl)-1-methylpyridinium methyl sulphate,
- [3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]trimethylammonium chloride,
- [3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]trimethylammonium methyl sulphate,
- 1-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-methylpiperidinium chloride,
- 1-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-methylpiperidinium methyl sulphate,
- 4-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-4-methylmorpholin-4-ium chloride,
- 4-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-4-methylmorpholin-4-ium methyl sulphate,

and their addition salts with an acid.

7. Compounds according to Claim 6, characterized in that they are selected from:

- 3-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-1-(2-hydroxyethyl)-3H-imidazol-1-ium chloride,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-methylpyridinium methyl sulphate,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-(2-hydroxyethyl)pyridinium chloride,
- 3-(3-amino-7-hydroxy-5-methylpyrazolo[1,5-a]-pyrimidin-6-ylmethyl)-1-methylpyridinium chloride,
- 4-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-4-methylmorpholin-4-ium chloride,
- 4-[3-(3-amino-5-methylpyrazolo[1,5-a]pyrimidin-7-ylamino)propyl]-4-methylmorpholin-4-ium methyl sulphate,

and their addition salts with an acid.

8. Compounds according to any one of the preceding claims, characterized in that the addition salts with an acid are selected from the hydrochlorides, hydrobromides, sulphates, citrates, succinates, tartrates, lactates and acetates.

9. Use of the compounds of formula (I) as defined in any one of the preceding claims as oxidation base precursors for the oxidation dyeing of keratinous fibres.

10. Composition for the oxidation dyeing of keratinous fibres, characterized in that it comprises,

as oxidation base, in a medium appropriate for dyeing, at least one compound of formula (I) as defined in any one of Claims 1 to 8.

11. Composition according to Claim 10, characterized in that the compound or compounds of formula (I) represent(s) from 0.0005 to 12% by weight of the total weight of the dyeing composition.

12. Composition according to Claim 11, characterized in that the compound or compounds of formula (I) represent(s) from 0.005 to 6% by weight of the total weight of the dyeing composition.

13. Composition according to any one of Claims 10 to 12, characterized in that it includes at least one additional oxidation base selected from para-phenylenediamines, bisphenylalkylenediamines, para-aminophenols, ortho-aminophenols and heterocyclic bases other than the compounds of formula (I).

14. Composition according to Claim 13, characterized in that the additional oxidation base or bases represent(s) from 0.0005 to 12% by weight of the total weight of the dyeing composition.

15. Composition according to any one of Claims 10 to 14, characterized in that it includes at least one coupler and/or at least one direct dye.

16. Composition according to Claim 15, characterized in that the coupler or couplers is or are selected from meta-phenylenediamines, meta-

aminophenols, meta-diphenols and heterocyclic couplers, and their addition salts with an acid.

17. Composition according to Claim 16, characterized in that the coupler or couplers is or are selected from 2-methyl-5-aminophenol, 5-N-(β-hydroxyethyl)amino-2-methylphenol, 3-aminophenol, 1,3-dihydroxybenzene, 1,3-dihydroxy-2-methylbenzene, 4-chloro-1,3-dihydroxybenzene, 2,4-diamino-1-(β-hydroxyethyloxy)benzene, 2-amino-4-(β-hydroxyethylamino)-1-methoxybenzene, 1,3-diaminobenzene, 1,3-bis(2,4-diaminophenoxy)propane, sesamol, α-naphthol, 6-hydroxyindole, 4-hydroxyindole, 4-hydroxy-N-methylindole, 6-hydroxyindoline, 2,6-dihydroxy-4-methylpyridine, 1H-3-methylpyrazol-5-one, 1-phenyl-3-methylpyrazol-5-one, and their addition salts with an acid.

18. Composition according to any one of Claims 15 to 17, characterized in that the coupler or couplers répresent(s) from 0.0001 to 10% by weight, approximately, of the total weight of the dyeing composition.

19. Composition according to any one of Claims 10 to 18, characterized in that the addition salts with an acid are selected from the hydrochlorides, hydrobromides, sulphates, citrates, succinates, tartrates, lactates and acetates.

20. Method of dyeing keratinous fibres and, in particular, human keratinous fibres such as the

hair, characterized in that at least one dyeing composition as defined in any one of Claims 10 to 19 is applied to the fibres, and in that the colour is revealed at an acidic, neutral or alkaline pH with the aid of an oxidizing agent which is added to the dyeing composition right at the time of use or which is present in an oxidizing composition which is applied simultaneously or sequentially, separately.

21. Method according to Claim 20, characterized in that the oxidizing agent is selected from hydrogen peroxide, urea peroxide, alkali metal bromates, persalts and enzymes.

22. Multi-compartment device or multi-compartment dyeing kit of which a first compartment contains a dyeing composition as defined in any one of Claims 10 to 19 and a second compartment contains an oxidizing composition.

PTO/SB/105 (8-96)
Approved for use through 9/30/98. OMB 0651-0032
Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

Attorney Docket No.: 05725.0944

Declaration and Power of Attorney for Patent Application

Déclaration et Pouvoir pour Demand de Brevet

French Language Declaration

En tant que l'inventeur nommé ci-après, je déclare par le présent acte que:

As a below named inventor, I hereby declare that:

Mon domicile, mon adresse postale et ma nationalité sont ceux figurant ci-dessous à côté de mon nom.

My residence, post office address and citizenship are as stated next to my name.

Je crois être le premier inventeur original et unique (si un seul nom est mentionné ci-dessous), ou l'un des premiers co-inventeurs originaux (si plusieurs noms sont mentionnés ci-dessous) de l'objet revendiqué, pour lequel une demande de brevet a été déposée concernant l'invention intitulée

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

NOVEL CATIONIC OXIDATION BASES, THEIR USE FOR THE OXIDATION DYEING OF KERATINOUS FIBRES, DYEING COMPOSITIONS AND METHODS OF DYEING

et dont la description est fournie ci-joint à moins que la case suivante n'ait été cochée:

the specification of which is attached hereto unless the following box is checked:

☒ a été déposée le _____
sous le numéro de demande des Etats-Unis ou le numéro de demande international PCT
_____ et modifiée
_____ (les cas échéant).

☒ was filed on January 14, 2000 as United States Application Number or PCT International Application Number PCT/FR00/00073 and was amended on _____ (if applicable).

Je déclare par le présent acte avoir passé en revue et compris le contenu de la description ci-dessus, revendications comprises, telles que modifées par toute modification dont il aura été fait référence ci-dessus.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above

Je reconnais devoir divulguer toute information pertinente à la brevetabilité, comme défini dans le Titre 37, § 1.56 du Code fédéral des réglementations.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56.

French Language Declaration **Attorney Docket No.:05725.0944**

Je revendique par le présent acte avoir la priorité étrangère, en vertu du Titre 35, § 119(a)-(d) ou § 365(b) du Code des Etats-Unis, sur toute demande étrangère de brevet ou certificat d'inventeur ou, en vertu du Titre 35, § 365(a) du même Code, sur toute demande internationale PCT désignant au moins un pays autre que les Etats-Unis et figurant ci-dessous et, en cochant la case, j'ai aussi indiqué ci-dessous toute demande étrangère de brevet, tout certificat d'inventeur ou toute demande internationale PCT ayant une date de dépôt précédant celle de la demande à propos de laquelle une priorité est revendiquée.

I hereby claim foreign priority under Title 35, United States Code, § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International Application which designated at least one country other than the United States, listed below, and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior foreign application(s)
Demande(s) de brevet antérieure(s)

Priority Not Claimed
Droit de priorité non revendiqué

(Number) (Numéro)	(Country) (Pays)	(Day/Month/Year Filed) (Jour/Mois/Anné de dépot)	☐
99/00503 ✓	France ✓	19 January 1999 ✓	
(Number) (Numéro)	(Country) (Pays)	(Day/Month/Year Filed) (Jour/Mois/Anné de dépot)	☐

Je revendique par le présent acte tout bénéfice, en vertu du Titre 35, § 119(c) du Code des Etats-Unis, de toute demande de brevet provisoire effectuée aux Etats-Unis et figurant ci-dessous.

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below.

(Application No.) (N° de demande)	(Filing Date) (Date de dépot)
(Application No.) (N° de demande)	(Filing Date) (Date de dépot)

Je revendique par le présent acte tout bénéfice, en vertu du Titre 35, § 120 du Code des Etats-Unis, de toute demande de brevet effectuée aux Etats-Unis, ou en vertu du Titre 35, § 365(c) du même Code, de toute demande internationale PCT désignant les Etats-Unis et figurant ci-dessous et, dans la mesure où l'objet de chacune des revendications de cette demande de brevet n'est pas divulgué dans la demande antérieure américaine ou internationale PCT, en vertu des dispositions du premier paragraphe du Titre 35, § 112 du Code des Etats-Unis, je reconnais devoir divulguer toute information pertinente à la brevetabilité, comme défini dans le Titre 37, § 1.56 du Code fédéral des réglementations, dont laquelle est devenue disponible entre la date de dépôt de la demande antérieure, et la date de dépôt de la demande nationale ou internationale PCT de la présente demande:

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International Application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International Application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose any or all information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

(Application No.) (N° de demande)	(Filing Date) (Date de dépot)	(Status) (patented, pending, abandoned) (Status) (breveté, en cours d'examen, abandonné)
(Application No.) (N° de demande)	(Filing Date) (Date de dépot)	(Status) (patented, pending, abandoned) (Status) (breveté, en cours d'examen, abandonné)

Je déclare par le présent acte que toute déclaration ci-incluse est, à ma connaissance, véridique et que toute déclaration formulée à partir de renseignements ou de suppositions est tenue pour véridique; et de plus, que toutes ces déclarations ont été formulées en sachant que toute fausse déclaration volontaire ou son équivalent est passible d'une amende ou d'une incarcération, ou des deux, en vertu de la Section 1001 du Titre 18 du Code des Etats-Unis, et que de telles déclarations volontairement fausses risquent de compromettre la validité de la demande de brevet ou du brevet délivré à partir de celle-ci.

I hereby declare that all statements made herei of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

French Language Declaration

Attorney Docket No.: 05725.0944

POUVOIRS: En tant que l'inventeur cité, je désigne par la présente l'(les) avocat(s) et/ou agent(s) suivant(s) pour qu'ils poursuive(nt) la procédure de cette demande de brevet et traite(nt) toute affaire s'y rapportant avec L'Office des brevets et des marques: *(mentionner le nom et le numéro d'enregistrement)*.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this patent application and transact all business in the Patent and Trademark Office connected therewith: (*list name and registration number*):

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	Attorney Docket No.: 05725.0944
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